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[Title of the Invention]

METHOD FOR PRODUCING PLASTIC LENSES, AND PLASTIC LENS

[Abstract]

[Problem] To solve the drawbacks of thiourethane-type plastic lenses and thioepoxy-type plastic lenses, and to obtain plastic lenses having a high refractive index and a high Abbe's number, and having good heat resistance, high mechanical strength and good colorability.

[Means for Resolution] A method for producing plastic lenses, which comprises mixing a polymerizing composition that contains the following components (A), (B) and (C), followed by polymerizing and curing it:

- (A) a compound having an episulfide group,
- (B) a compound having a mercapto group,
- (C) a compound having an isocyanate or isothiocyanate group.

[Claims]

1. A method for producing plastic lenses, which

comprises mixing a polymerizing composition that contains, as the essential ingredients, at least the following three components (A), (B) and (C), followed by polymerizing and curing the polymerizing composition:

- (A) a compound having an episulfide group,
- (B) a compound having a mercapto group,
- (C) a compound having an isocyanate or isothiocyanate group.

2. The method for producing plastic lenses as claimed in claim 1, wherein the blend ratio of the components (A), (B) and (C) is as follows:

- (A): from 5 to 95 parts by weight,
- (B) and (C) in total: from 5 to 95 parts by weight.

3. The method for producing plastic lenses as claimed in claim 1 or 2, wherein the molar number of the mercapto group in (B),  $B_m$ , and the molar number of the isocyanate or isothiocyanate group in (C),  $C_m$ , are within a range of  $0.5 < C_m/B_m < 2$ .

4. A plastic lens produced according to the method of any one of claims 1 to 3.

5. The plastic lens as claimed in claim 4, which is coated with a hard coat layer.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Belongs]

The present invention relates to a method for producing plastic lenses such as those for eyesight correction, those for sunglasses, those for fashion glasses, those for photochromic lenses, those for cameras, those for optical devices, etc.

[0002]

[Prior Art]

Various plastic lenses have heretofore been studied, which could be substitutes for conventional inorganic glass lenses for optical use. As being lightweight, resistant to shock, and colorable with ease, plastic lenses are much used for spectacles these days. For lenses for spectacles, in general, diethylene glycol bisallylcarbonate (CR-39) is widely used. However, as its refractivity is low, CR-39 is often problematic in that it could not be formed into thin and lightweight lenses. Therefore, for optical use, for example, for spectacles, various materials and methods have heretofore been proposed for producing high-refractivity lenses. However, high-refractivity lenses are often problematic in that their Abbe's number is generally low. To reduce their chromatic aberration, lenses must have a high Abbe's number.

[0003]

In Japanese Patent Publication No. 58489/1997 and Japanese Patent Laid-Open No. 148340/1993, proposed are

thermosetting plastic lenses having a thiourethane structure formed through reaction of a polythiol compound and a polyisocyanate compound. In Japanese Patent Laid-Open Nos. 71580/1997, 110979/1997 and 255781/1997, proposed are thioepoxy-type plastic lenses formed by polymerizing and curing an episulfide compound.

[0004]

[Problems that the Invention is to Solve]

The thiourethane-type plastic lenses have the advantages of high refractivity and high strength at break, but have the disadvantage of poor heat resistance. The thioepoxy-type plastic lenses have a high refractive index and a high Abbe's number and are resistant to heat, but have the disadvantage of low strength at break. Especially for use in spectacles, the lenses are problematic in that they are brittle. In addition, the thioepoxy-type plastic lenses have an extremely low water absorption, and are therefore further problematic in that they take a lot of time for coloration. In fact, they are difficult to color for use in spectacles.

[0005]

[Means for Solving the Problems]

We, the present inventors have assiduously studied so as to solve the problems noted above, and, as a result, have found that the problems can be solved by polymerizing and

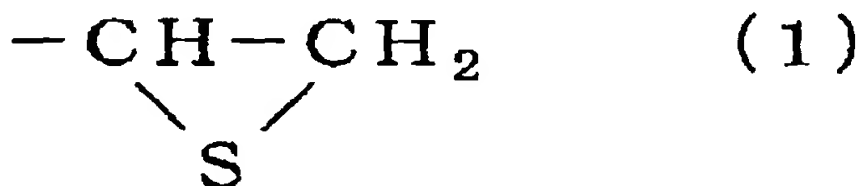
curing a composition that contains polymerizing monomers (A), (B) and (C) mentioned below. On the basis of this finding, we have completed the present invention.

[0006]

- (A) a compound having an episulfide group,
- (B) a compound having a mercapto group,
- (C) a compound having an isocyanate or isothiocyanate group.

The episulfide group-containing compound is meant to indicate a compound that contains at least one functional group of the following formula (1) in one molecule.

[0007]



[0008]

Thiourethane-type plastic lenses have the advantages of high refractivity and high mechanical strength but have the disadvantage of poor heat resistance. This is because, for high-refractivity plastic lenses produced through reaction of a polythiol and a polyisocyanate, the crosslinking density of the polymer is difficult to increase

as interfering with the refractivity of the lenses. On the other hand, thioepoxy-type plastic lenses have a high refractive index and a high Abbe's number and are resistant to heat, but have the disadvantage of low mechanical strength and poor colorability. This is because, for thioepoxy-type plastic lenses produced through reaction of the episulfide groups of the monomer, the crosslinking density of the polymer inevitably increases to a great extent, and, as a result, the lenses could not be tough and their mechanical strength is thereby lowered. In addition, in general, thioepoxy-type plastic lenses hardly absorb water and are resistant to heat. Therefore, the lenses are further problematic in that they take a lot of time for coloration.

[0009]

In producing plastic lenses, it is a matter of great importance to control the crosslinking density of polymers. However, in producing thiourethane-type plastic lenses, it is difficult to increase the crosslinking density of the polymer formed through reaction of only a polythiol and a polyisocyanate with the high refractivity of the polymer being kept as it is. On the other hand, in producing thioepoxy-type plastic lenses to have a lowered crosslinking density through reaction of only the episulfide groups of the monomer, the episulfide group-having monomer compound must be a bi-functional, long single-chain polymerizing monomer.

However, this is not practicable in view of the probably complicated process of producing the starting monomer.

[0010]

In the present invention, the three compounds, one having an episulfide group, another having a mercapto group and still another having an isocyanate or isothiocyanate group, are first mixed, and then polymerized and cured. In the process, the crosslinking density of the polymer produced can be well controlled to a desired one, and, as a result, the plastic lens of the polymer can have an increased refractive index and an increased Abbe's number and can have good heat resistance and high strength at break. In addition, the water absorption of the lens of the polymer is higher in some degree than that of thioepoxy-type lenses. Therefore, the lens of the invention can be readily colored.

[0011]

The blend ratio of the components (A), (B) and (C) to form the composition for use in the invention is not specifically defined, but is preferably as follows:

(A): from 5 to 95 parts by weight, and

(B) and (C) in total: from 5 to 95 parts by weight.

If the amount of the component (A) is smaller than 5 parts by weight and that of the components (B) and (C) in total is larger than 95 parts by weight, the crosslinking density of the polymer formed could not increase to a satisfactory

degree, like that for thiourethane-type plastic lenses. If so, the heat resistance of the plastic lenses made from the polymer will be low. On the other hand, if the amount of the components (B) and (C) in total is smaller than 5 parts by weight and that of the component (A) is larger than 95 parts by weight, the crosslinking density of the polymer formed will be too high, like that for thioepoxy-type plastic lenses. If so, the strength at break of the plastic lenses made from the polymer will be low, or that is, the lenses will be extremely brittle.

[0012]

In case where the amount of the mercapto group, the isocyanate group or the isothiocyanate group remaining in the plastic lenses produced by polymerizing and curing the monomer composition is too large, the weather resistance of the lenses will be poor. Therefore, it is desirable to reduce the amount of the mercapto group, the isocyanate group or the isothiocyanate group remaining in the plastic lenses produced. For this, the blend ratio of the components (B) and (C) is preferably so controlled that the molar number of the mercapto group in (B),  $B_m$ , and the molar number of the isocyanate or isothiocyanate group in (C),  $C_m$ , are within a range of  $0.5 < C_m/B_m < 2$ .

[0013]

The episulfide group-containing compound for use in



the invention is not specifically defined, and any and every known compound having an episulfide group is usable herein with no limitation. For example, the epoxy group of a known epoxy compound may be partly or entirely episulfidized to give an episulfide compound for use in the invention. For further increasing the refractive index and the Abbe's number of the plastic lenses of the invention, it is more desirable that the episulfide compound for the lenses contains sulfur atom(s) in addition to the episulfide group. Concretely, the episulfide compound of the type includes 1,2-bis( $\beta$ -epithiopropylthio)ethane, bis( $\beta$ -epithiopropyl) sulfide, 1,4-bis( $\beta$ -epithiopropylthiomethyl)benzene, and 2,5-bis( $\beta$ -epithiopropylthiomethyl)-1,4-dithian. The mercapto group-containing compound for use in the invention is not also specifically defined, and any and every known compound having a mercapto group is usable herein with no limitation. Specific examples of known, mercapto group-containing compounds usable in the invention are aliphatic polythiols such as 1,2-ethanedithiol, 1,6-hexanedithiol, 1,1-cyclohexanedithiol, etc.; and aromatic polythiols such as 1,2-dimercaptobenzene, 1,2,3-tris(mercaptomethyl)benzene, etc. For further increasing the refractive index and the Abbe's number of the plastic lenses of the invention, it is more desirable that the mercapto compound for the lenses contains sulfur atom(s) in addition to the mercapto group.

Concretely, the mercapto compound of the type includes 1,2-bis(mercaptomethylthio)benzene, 1,2,3-tris(mercaptoethylthio)benzene, and 1,2-bis((2-mercaptoethyl)thio)-3-mercaptopropane.

[0014]

The isocyanate or isothiocyanate group-containing compound for use in the invention is not also specifically defined, and any and every known, isocyanate or isothiocyanate group-containing compound is usable herein with no limitation. Specific examples of the isocyanate group-containing compound are ethylene diisocyanate, trimethylene diisocyanate, 2,4,4-trimethylhexane diisocyanate, hexamethylene diisocyanate, and m-xylylene diisocyanate.

[0015]

The polymerizing composition prepared by mixing the three components (A), (B) and (C) may be polymerized and cured in the presence of at least one curing catalyst to give plastic lenses. The curing catalyst is not specifically defined, and may be any and every ordinary one for epoxy resins or urethane resins. Specific examples of the curing catalyst are amine compounds such as ethylamine, ethylenediamine, triethylamine, tributylamine, etc.; as well as dibutyltin dichloride, dimethyltin dichloride, etc.

[0016]

Before being polymerized and cured, the polymerizing composition of the invention may be optionally mixed with a UV stabilizer, and the resulting mixture may be polymerized and cured to give plastic lenses. The lenses thus produced may have further improved weather resistance. Specific examples of the UV stabilizer are hindered amine-type light stabilizers, hindered phenol-type antioxidants, phosphite-type antioxidants, thioether-type antioxidants, benzotriazole-type UV absorbents, and benzophenone-type UV absorbents.

[0017]

When the plastic photochromic lenses of the invention are used for eyesight correction or for fashion glasses, it is desirable that the lenses are coated with an anti-glare film capable of increasing the light transmittance through the lenses and of preventing surface reflection to cause the glare on the lenses. In addition, it is also desirable that the lenses are coated with a hard coat layer for increasing the adhesiveness between the lens substrate and the anti-glare film and for preventing the lens surface from being scratched.

[0018]

One preferred example for the hard coat layer is a coating composition that comprises the following essential components (a) and (b). After applied to the lenses, the

composition is cured to form a hard coat layer.

(a) At least one silane compound having at least one reactive group.

(b) At least one type selected from metallic particles of silicon oxide, antimony oxide, zirconium oxide, titanium oxide, tin oxide, tantalum oxide, tungsten oxide, aluminium oxide or the like; composite metallic particles of at least two of titanium oxide, cerium oxide, zirconium oxide, silicon oxide and iron oxide; and composite metallic particles prepared by coating tin oxide particles with composite metallic particles of tin oxide and tungsten oxide.

[0019]

The component (b) is effective for controlling the refractivity of the hard coat layer and for increasing the hardness of the layer. One or more types of the metallic particles for the component (b) may be used either singly or as combined. However, the component (b) alone could not form a good film, and, when combined with the component (a), it can form a transparent and tough film. The component (a) may be directly used as it is, but is preferably hydrolyzed prior to being combined with the component (b). After hydrolyzed, the component (a) is more effective for increasing the water resistance and the hardness of the hard coat film formed.

[0020]

In general, the thickness of the hard coat layer

preferably falls between 0.2  $\mu\text{m}$  and 10  $\mu\text{m}$  or so, more preferably between 1  $\mu\text{m}$  and 3  $\mu\text{m}$  or so. If desired, a primer layer may be provided between the lens substrate and the hard coat layer. The primer layer enhances the adhesiveness between the lens substrate and the hard coat layer, and enhances the impact resistance of lenses thus coated with the hard coat layer.

[0021]

For eyesight correction, the lenses coated with the hard coat layer are preferably further coated with an anti-glare film, as so mentioned hereinabove. Being thus coated, the lenses have further improved optical properties. The anti-glare film may be a multi-layered film to be prepared by laminating a plurality of thin films each having a different refractive index. For this, any inorganic or organic substances are usable, provided that they can reduce the refractivity of lenses. However, essentially for increasing the surface hardness of the lenses and for removing the interference fringes from the lenses, most preferred is a single-layered or multi-layered, inorganic anti-glare film. The inorganic substance usable for the film includes oxides and fluorides such as silicon oxide, aluminium oxide, zirconium oxide, titanium oxide, cerium oxide, hafnium oxide, magnesium fluoride, etc. The film may be formed on the lenses through PVD such as ion plating, vacuum vapor deposition,

sputtering, etc.

[0022]

[Modes of Carrying out the Invention]

The invention is described in detail with reference to the following Examples, which, however, are not intended to restrict the scope of the invention. The details of the abbreviations of the substances used in Examples and Comparative Examples are mentioned below.

[0023]

Episulfide group-containing compounds:

Abbreviation: Name of the substance

A-1: bis( $\beta$ -epithiopropyl) sulfide

A-2: 1,4-bis( $\beta$ -epithiopropylthiomethyl)benzene

A-3: 2,5-bis( $\beta$ -epithiopropylthiomethyl)-1,4-dithian

Mercapto group-containing compound:

Abbreviation: Name of the substance

B-1: 1,2-bis((2-mercaptoethyl)thio)-3-mercaptopropane

Isocyanate or isothiocyanate group-containing compounds:

Abbreviation: Name of the substance

C-1: m-xylylene diisocyanate

C-2: 1,4-bis(cyclohexane isocyanate)

(Example 1)

The components shown in Table 1 were mixed to prepare a composition (100 g) for plastic lenses. To this were added 0.05 g of a UV absorbent, SEESORB701 (from Shipro Chemical),

and 0.05 g of a polymerization catalyst, dibutyltin dichloride, and well stirred at room temperature. Then, the resulting mixture was further stirred under a reduced pressure of 5 mmHg to degas it for 30 minutes. This was cast into a mirror-finished glass mold equipped with a gasket, heated therein from 35°C to 120°C over a period of 16 hours. After having been thus polymerized and cured, the lens formed was taken out of the mold, and then annealed under heat at 120°C for 2 hours.

[0024]

The plastic lens thus formed was evaluated according to the methods mentioned below. Its data are given in Table 2.

Refractive Index:

Measured with an Abbe's refractometer, the refractive index is for D rays of 589.3 nm at 20°C.

Abbe's Number:

Measured with an Abbe's refractometer, the Abbe's number is at 20°C.

Heat Resistance:

Measured with a TMA tester under a load of 50 g, Tg indicates the heat resistance of the lens.

Flexural Test:

The flexural strength of the lens is measured according to JIS K7203.

[0025]

Coloration:

A thermostat chamber filled with water and controlled at 90°C was prepared. A glass beaker filled with one liter of water was put in the chamber, and this was used as a dyeing pot. 1.5 g of a colorant, Seiko Placs Gray (from Hattori Seiko), 3 cc of a surfactant serving as a dispersant, NES-203 (from Nikko Chemicals), and 10 cc of a colorant carrier, benzyl alcohol were added to the pot, and well stirred and mixed to prepare a dyeing bath. The plastic lens formed was completely sunk in the dyeing bath. After left as such for 20 minutes, the lens was taken out of the bath. The visible ray transmittance through the thus-colored lens was measured with a BPI photometer, and this indicates the color density of the lens. (Lenses having a lower visible ray transmittance are colored to a higher degree.)



[ 0026 ]

[ Table 1 ]

	Component A (g)	Component B (g)	Component C (g)
Example 1	A-1: 60 g	B-1: 20 g	C-1: 20 g
Example 2	A-1: 50 g	B-1: 25 g	C-2: 25 g
Example 3	A-2: 60 g	B-1: 20 g	C-1: 20 g
Example 4	A-3: 60 g	B-1: 20 g	C-1: 20 g
Example 5	A-3: 60 g	B-1: 20 g	C-2: 20 g
Comparative Example 1	A-1: 100 g	-	-
Comparative Example 2	A-2: 100 g	-	-
Comparative Example 3	A-1: 98 g	B-1: 1 g	C-1: 1 g
Comparative Example 4	-	B-1: 50 g	C-1: 50 g
Comparative Example 5	-	B-1: 50 g	C-2: 50 g

[ 0027 ]

[ Table 2 ]

	Refractive Index ( $n_D$ )	Abbe's Number ( $v_D$ )	Heat Resistance T <sub>g</sub> (°C)	Flexural Strength (kg/cm <sup>2</sup> )	Visible Ray Transmittance through Colored Lens (%)
Example 1	1.70	34	94	1950	61
Example 2	1.68	32	91	1980	57
Example 3	1.70	32	93	1880	60
Example 4	1.71	34	90	2000	58
Example 5	1.69	35	96	1800	60
Comparative Example 1	1.70	35	softening point not appeared	1200	87
Comparative Example 2	1.67	34	softening point not appeared	1400	86
Comparative Example 3	1.70	35	softening point not appeared	1240	84
Comparative Example 4	1.66	33	81	2200	34
Comparative Example 5	1.62	39	120	2100	46

[0028]

(Examples 2 to 5)

Plastic lenses were produced in the same manner as in Example 1, for which, however, used were the components shown in Table 1 to prepare the starting composition (100 g). These were tested also in the same manner as in Example 1, and their data are given in Table 2.

[0029]

(Comparative Examples 1 and 2)

Plastic lenses were produced in the same manner as in Example 1, for which, however, used were the components shown in Table 1 to prepare the starting composition (100 g). These were tested also in the same manner as in Example 1, and their data are given in Table 2.

[0030]

(Comparative Examples 3 to 5)

Plastic lenses were produced in the same manner as in Example 1, for which, however, used were the components shown in Table 1 to prepare the starting composition (100 g). The polymerization catalyst for the compositions was tributylamine (0.05 g). These were tested also in the same manner as in Example 1, and their data are given in Table 2.

[0031]

[Advantages of the Invention]

The method of the invention for producing plastic lenses

solves the drawbacks of thiourethane-type plastic lenses and thioepoxy-type plastic lenses. The plastic lenses produced in the method have a high refractive index and a high Abbe's number, and have increased heat resistance and increased mechanical strength, and they are easy to color.

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(54) 【発明の名称】 プラスチックレンズの製造方法及びプラスチックレンズ

(57) 【要約】

【課題】 チオウレタン系プラスチックレンズとチオエポキシ系プラスチックレンズの欠点を改良し、高屈折率、高アッペ数で、耐熱性が高く、機械的強度に優れかつ染色性も良好な、プラスチックレンズを得る。

【解決手段】 下記(A)、(B)、(C)の成分を含む重合性組成物を混合した後、重合硬化することによって製造する、プラスチックレンズの製造方法。

(A) エピスルフィド基を含む化合物

(B) メルカプト基を含む化合物

(C) イソシアナート基またはイソチオシアナート基を含む化合物

## 【特許請求の範囲】

【請求項1】 少なくとも下記(A)、(B)、(C)の3成分を主成分とする重合性組成物を混合した後、該重合性組成物を重合硬化させて製造することを特徴とする、プラスチックレンズの製造方法。

(A) エピスルフィド基を含む化合物

(B) メルカプト基を含む化合物

(C) イソシアナート基またはイソチオシアナート基を含む化合物

【請求項2】 請求項1に記載のプラスチックレンズの製造方法において、前記(A)、(B)、(C)の組成比が、以下に示す範囲であることを特徴とするプラスチックレンズの製造方法。

(A) : 5~95重量部、

(B)と(C)の合計: 5~95重量部

【請求項3】 請求項1または2に記載のプラスチックレンズの製造方法において前記(B)のメルカプト基のモル数をBm、前記(C)のイソシアナート基またはイソチオシアナート基のモル数をCmとしたときに、 $0.5 < Cm/Bm < 2$ の範囲にあることを特徴とするプラスチックレンズの製造方法。

【請求項4】 請求項1ないし3のいずれか1項に記載の製造方法で製造されたプラスチックレンズ。

【請求項5】 前記プラスチックレンズ表面にハードコート層が設けられていることを特徴とする、請求項4記載のプラスチックレンズ。

## 【発明の詳細な説明】

## 【0001】

【発明の属する技術分野】 本発明は、矯正用レンズ、サングラスレンズ、ファッションレンズ、フォトクロミックレンズ、カメラ用レンズ、光学装置用レンズ等に用いられるプラスチックレンズの製造方法に関する。

## 【0002】

【従来の技術】 従来、光学用途に用いられてきた無機ガラスレンズに代わる物として、以前より種々のプラスチックレンズが研究されている。プラスチックレンズは軽量で耐衝撃性に優れ、かつ染色が容易であることから、眼鏡レンズ用として近年多用されている。一般に眼鏡レンズ等には、ジェチレングリコールビスアリルカーボネート(CR-39)が幅広く用いられているが、レンズを薄型化、軽量化する上で、CR-39の低い屈折率が問題となっている。このため眼鏡レンズ等の光学用途に対して、高屈折率のレンズを作製するために、様々な材料や製造方法が提案されている。また、高屈折率素材では一般にアッペ数が低くなる傾向があるが、レンズの色収差を低減するために高アッペ数化も必要になっている。

【0003】 特公平4-58489号公報、特開平5-148340号公報には、ポリチオール化合物とポリイソシアナート化合物との反応によるチオウレタン構造を

持つ熱硬化型プラスチックレンズが提案されている。また、特開平9-71580号公報、特開平9-110979号公報、特開平9-255781号公報にはエピスルフィド化合物の重合硬化によるチオエポキシ系のプラスチックレンズが提案されている。

## 【0004】

【発明が解決しようとする課題】 これらのチオウレタン系プラスチックレンズは、高屈折率で、破壊強度が強いという長所を持つが、耐熱性が低いという短所があった。また、チオエポキシ系プラスチックレンズは、高屈折率、高アッペ数であり、耐熱性に優れるという長所を持つが、破壊強度が弱いという短所があり、特に眼鏡レンズ用途にはレンズ素材としての脆さが問題となっている。また、チオエポキシ系プラスチックレンズはレンズ素材の吸水率が極めて低いため、染色加工時間が長時間となり、眼鏡レンズ用としての染色加工は困難であった。

## 【0005】

【課題を解決するための手段】 本発明者らは、かかる問題点を解決するべく鋭意研究を重ねた結果、下記(A)、(B)、(C)の重合性単量体を含む化合物を、重合硬化することによって、上記問題点が解決できることを見だし、本発明を完成するに至った。

## 【0006】 (A) エピスルフィド基を含む化合物

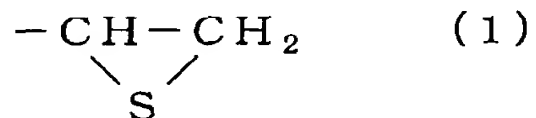
## (B) メルカプト基を含む化合物

## (C) イソシアナート基またはイソチオシアナート基を含む化合物

なお、エピスルフィド基を含む化合物とは、下記一般式(1)で示される官能基を1分子中に1つ以上含む化合物を意味する。

## 【0007】

## 【化1】



【0008】 チオウレタン系プラスチックレンズは、高屈折率で、機械強度が強いという長所を持つが、耐熱性が低いという短所があった。これはポリチオールとポリイソシアナートの反応によって高屈折率のプラスチックレンズを製造するにあたっては、高屈折率化の為に、ポリマーの架橋密度を上げることが困難なためである。また、チオエポキシ系プラスチックレンズは、高屈折率、高アッペ数であり、耐熱性に優れるという長所を持つが、機械強度が弱く、染色性に劣るという短所があった。これはエピスルフィド基同士の反応によって、チオエポキシ系プラスチックレンズを製造するにあたっては、ポリマー中の架橋密度が非常に高くなることが避けられず、その結果レンズの靱性がなくなることによって機械強度が低下するためである。また、チオエポキシ系